

Full counting statistics of spin transfer through ultrasmall quantum dots

T. L. Schmidt,¹ A. Komnik,^{1,2} and A. O. Gogolin³

¹*Physikalisches Institut, Universität Freiburg, D-79104 Freiburg, Germany*

²*Institut für Theoretische Physik, Universität Heidelberg, D-69120 Heidelberg, Germany*

³*Department of Mathematics, 180 Queen's Gate, London SW7 2AZ, United Kingdom*

(Dated: February 1, 2008)

We analyze the spin-resolved full counting statistics of electron transfer through an ultrasmall quantum dot coupled to metallic electrodes. Modelling the setup by the Anderson Hamiltonian, we explicitly take into account the onsite Coulomb repulsion U . We calculate the cumulant generating function for the probability to transfer a certain number of electrons with a *preselected* spin orientation during a fixed time interval. With the cumulant generating function at hand we are then able to calculate the spin current correlations which are of outmost importance in the emerging field of spintronics. We confirm the existing results for the charge statistics and report the discovery of the new type of correlation between the spin-up and -down polarized electrons flows, which has a potential to become a powerful new instrument for the investigation of the Kondo effect in nanostructures.

PACS numbers: 72.10.Fk, 72.25.Mk, 73.63.-b

Modern microelectronics is one of the most successful technologies ever conceived by humankind. However, in recent years a number of limitations, which can slow down or even stop further progress began to come to the fore. One possible way to overcome these difficulties is to switch from charge current processing to spin current and spin configuration processing. Their advantages are so enormous that recently a completely new scientific field of spintronics has been established [1, 2].

In the conventional microelectronics the properties of the basic circuitry elements are characterized by a number of different quantities – by the nonlinear current-voltage relations, by the current noise spectra, by the current correlations of third order (third cumulant) etc [3, 4]. However, there is one characteristic which (at least in the low frequency range) contains information about correlations of *all* orders. This is the so-called full counting statistics (FCS), which answers all above questions by providing the probability distribution $P(Q)$ to transfer a certain amount of charge Q during a waiting time interval \mathcal{T} [5, 6].

While there is by now a vast amount of literature available on the charge transfer statistics, its spin-resolved relative remains to a larger part unknown with some notable exceptions [7, 8, 9, 10]. We would like to close this gap and analyze the combined statistics of spin and charge transfer through ultrasmall quantum dots with genuine repulsive interactions. The distinctive feature of these devices are their extremely small lateral dimensions which allow for only very few energy levels to take part in the transport processes. Typical realizations are nanoscale heterostructures on the semiconductor basis, or individual molecules coupled to metallic electrodes, which are even smaller [11, 12, 13, 14]. Both types of systems became available only during the last decades and are promising candidates to become basic building

blocks of future nanoelectronics and spintronics circuitry.

The archetype system to describe such structures is the single-impurity Anderson model [15, 16]. It consists of a local fermionic level (also called dot level) which is filled or emptied by $d_\sigma^\dagger, d_\sigma$ creation and annihilation operators and N different electronic continua modeling the electrodes via fermionic fields $\psi_{\alpha\sigma}(x)$,

$$H_0 = \sum_{\sigma=\pm} (\Delta + \sigma h) d_\sigma^\dagger d_\sigma + \sum_{\alpha=1}^N \sum_{\sigma=\pm} H_0[\psi_{\alpha\sigma}], \quad (1)$$

where $\Delta + \sigma h =: \delta_\sigma$ is responsible for the energy of the dot in the magnetic field $h = g\mu_B B/2$, when it is occupied by an electron with spin orientation σ . The dot level energy Δ is an additional parameter which can in practice be changed by varying the voltage on the background gate electrode. The coupling of dot and leads is achieved by a local tunneling contribution

$$H_T = \sum_{\alpha} \sum_{\sigma} \gamma_{\alpha} [d_\sigma^\dagger \psi_{\alpha\sigma}(x=0) + \text{h.c.}] , \quad (2)$$

with energy independent tunneling amplitudes γ_{α} . In addition to these terms one has to take into account the electrostatic repulsion

$$H_U = U(d_\uparrow^\dagger d_\uparrow - n_0)(d_\downarrow^\dagger d_\downarrow - n_0) , \quad (3)$$

reflecting the energetic cost U of double dot occupation with respect to the symmetric value $n_0 = 1/2$. Due to this normalization, the particle-hole symmetric case corresponds to $\Delta = 0$. The full system Hamiltonian is the sum of all three contributions $H = H_0 + H_T + H_U$.

The technology for the calculation of the FCS is by now far advanced and allows for a number of different approaches. In the most widespread one, the quantity of interest is the so-called cumulant generating function

(CGF) $\ln \chi(\lambda_\downarrow, \lambda_\uparrow) = \ln \chi(\lambda_\sigma)$ [6, 17, 18]. Its successive differentiation with respect to the counting fields λ_σ yields the respective irreducible momenta $\langle\langle \delta Q_\sigma^n \rangle\rangle$ for the probability distribution to transfer δQ_σ charges with spin orientation σ through the system during the waiting time \mathcal{T} ,

$$\langle\langle \delta Q_\sigma^n \rangle\rangle = (-i)^n \frac{\partial^n}{\partial \lambda_\sigma^n} \ln \chi(\lambda_\sigma) \Big|_{\lambda=0}. \quad (4)$$

In analogy to the approaches taken in [19, 20], the first step in the calculation of the CGF is to endow the tunneling Hamiltonian (2) with counting fields λ_σ . Outside of the waiting time interval $0 < t < \mathcal{T}$ the counting fields are zero. The tunneling Hamiltonian then transforms to

$$H_T \rightarrow H_T^\lambda = \sum_\alpha \sum_\sigma \gamma_\alpha \left[e^{i\lambda_\alpha \sigma / 2} d_\sigma^\dagger \psi_{\alpha\sigma} + \text{h.c.} \right]. \quad (5)$$

In the noninteracting case ($U = 0$, resonant level model [20, 21]) this quantity can easily be calculated by resummation of the perturbation series in γ_α or by applying the Levitov-Lesovik formula [5]. Assuming the measurement time \mathcal{T} to be large such that switching effects can be neglected, one finds (setting $e = \hbar = 1$)

$$\begin{aligned} \ln \chi_0(\lambda_\sigma) &= \mathcal{T} \sum_\sigma \int \frac{d\omega}{2\pi} \ln \left\{ 1 + \sum_{\alpha\beta} T_\sigma^{\alpha\beta}(\omega) n_\alpha (1 - n_\beta) \right. \\ &\times \left. [e^{i(\lambda_{\alpha\sigma} - \lambda_{\beta\sigma})} - 1] \right\}, \end{aligned} \quad (6)$$

where $n_\beta(\omega)$ denotes the Fermi distribution in lead β . The energy dependent transmission coefficients are given by

$$T_\sigma^{\alpha\beta}(\omega) = \frac{4\Gamma_\alpha\Gamma_\beta}{(\omega - \delta_\sigma)^2 + \Gamma^2}, \quad (7)$$

where $\Gamma_\beta = \pi\nu\gamma_\beta^2$ (with the density of states ν at the Fermi level) is the hybridization of the dot with lead β and $\Gamma = \sum_\beta \Gamma_\beta$.

While the non-interacting result (6) was derived for an arbitrary number N of fermionic leads, we shall restrict ourselves henceforth to the symmetric two level case, i. e., we assume $N = 2$ and $\Gamma_L = \Gamma_R$. The chemical potentials of the leads are assumed to be at $\mu_{L,R}$ where $V = \mu_L - \mu_R$ denotes the applied voltage.

It is quite inefficient to calculate $\chi(\lambda_\sigma)$ using an additional expansion in U in the same way. As has been realized in Ref. [22] in a different context, as long as one is only interested in small U , the CGF can be calculated by a simple linked cluster like calculation. Thereby the

full λ_σ dependence can be shifted onto the unperturbed Keldysh Green's functions $D(\omega)$ of the dot level. Using the notation of [23], these are given by

$$\begin{aligned} D^{--}(\omega) &= \left[(\omega - \delta_\sigma) + \sum 2i\Gamma_\beta(n_\beta - 1/2) \right] / \mathcal{D}(\omega), \\ D^{-+}(\omega) &= \left[\sum 2i\Gamma_\beta e^{i\lambda_{\beta\sigma}} n_\beta \right] / \mathcal{D}(\omega), \\ D^{+-}(\omega) &= \left[\sum 2i\Gamma_\beta e^{-i\lambda_{\beta\sigma}} (n_\beta - 1) \right] / \mathcal{D}(\omega), \\ D^{++}(\omega) &= -[D^{--}(\omega)]^*, \end{aligned} \quad (8)$$

where we defined

$$\begin{aligned} \mathcal{D}(\omega) &= (\omega - \delta_\sigma)^2 + \Gamma^2 \\ &+ 4 \sum_{\alpha\beta} \Gamma_\alpha \Gamma_\beta n_\alpha (1 - n_\beta) \left[e^{i(\lambda_{\alpha\sigma} - \lambda_{\beta\sigma})} - 1 \right]. \end{aligned} \quad (9)$$

Note that the presence of the counting field causes a violation of Keldysh's sum rule. Using a linked cluster expansion, the exact CGF $\ln \chi(\lambda_\sigma)$ can be expressed as a correction to the noninteracting CGF (6). The quantity we have to evaluate is then

$$\chi(\lambda_\sigma) = \chi_0(\lambda_\sigma) \langle T_C \exp \left[-i \int_C dt H_U(t) \right] \rangle. \quad (10)$$

The expectation value is to be taken with respect to the noninteracting ground state and therefore contains the λ -dependent Green's functions (8). It may appear that since the lowest order expansion in U only contains the dot occupation numbers n_σ , one is not expecting any counting field dependence to survive. However, this is no longer valid in the case of explicitly (quite artificially) time-dependent λ_σ . In the limit

$$\max\{\Delta, h, V\}/\Gamma \ll 1, \quad (11)$$

the first order contribution is given by

$$\ln \chi^{(1)}(\lambda_\sigma) = -\frac{UTV}{\pi^2\Gamma^3} \sum_\sigma \delta_\sigma \delta_{\bar{\sigma}} (e^{-i\lambda_\sigma} - 1), \quad (12)$$

where $\bar{\sigma} = -\sigma$. As it depends on δ_σ , this term only contributes for finite magnetic field ($h \neq 0$) and/or broken electron-hole symmetry ($\Delta \neq 0$).

The second order contribution is given by two different diagrams, see Fig. 1. One of these is again proportional to the magnetic field and contains the average dot occupation numbers while the other one is the double shell diagram. The calculation procedure is rather lengthy but straightforward and results in the following CGF expansion in the limit (11),

$$\begin{aligned} \ln \chi^{(2)}(\lambda_\sigma) &= \frac{\mathcal{T}V\chi_o^2}{2\pi\Gamma^2} \sum_\sigma \delta_\sigma^2(e^{-i\lambda_\sigma} - 1) + \frac{\mathcal{T}V(\chi_e^2 - 1)}{2\pi\Gamma^2} \sum_\sigma \delta_\sigma^2(e^{-i\lambda_\sigma} - 1) \\ &+ \frac{\mathcal{T}V^3\chi_o^2}{24\pi\Gamma^2} \left\{ 4(e^{-i\lambda_\uparrow - i\lambda_\downarrow} - 1) + \sum_\sigma (e^{-i\lambda_\sigma} - 1) \right\} + \frac{\mathcal{T}V^3(\chi_e^2 - 1)}{24\pi\Gamma^2} \sum_\sigma (e^{-i\lambda_\sigma} - 1), \end{aligned} \quad (13)$$

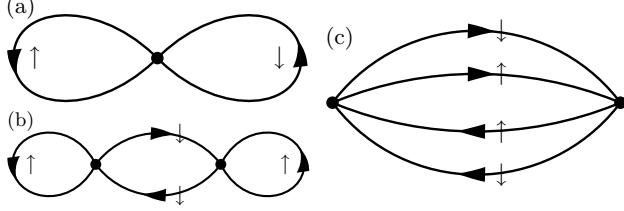


FIG. 1: One first order (a) and two second order (b,c) diagrams.

where we introduced the equilibrium even/odd susceptibilities (correlations of n_\uparrow with n_\uparrow and n_\downarrow , respectively), which are known to possess the following expansions for small U [24, 25, 26],

$$\chi_e = 1 + \left(3 - \frac{\pi^2}{4}\right) \frac{U^2}{\pi^2\Gamma^2}, \quad \chi_o = -\frac{U}{\pi\Gamma}. \quad (14)$$

Following the reasoning along the lines of Ref. [20], we may speculate that (13) is the exact result to all orders of U and Γ as soon as one inserts the exact values for $\chi_{e,o}$ which have been obtained by, e.g., Bethe ansatz calculations [27, 28]. Now we are in a position to establish contact to known results and to discuss new effects. Thus far, similar results have been obtained only for the charge transport statistics of the same system for the much more restrictive particle-hole symmetric parameter constellation [20]. The complete spin resolved statistics for large transmission Γ is given by the following CGF,

$$\begin{aligned} \ln \chi(\lambda_\sigma) &= iG_0\mathcal{T}V(\lambda_\uparrow + \lambda_\downarrow) + \frac{\mathcal{T}V}{2\pi\Gamma^2} \sum_\sigma (\chi_e\delta_\sigma + \chi_o\delta_\sigma)^2(e^{-i\lambda_\sigma} - 1) \\ &+ \frac{\chi_o^2\mathcal{T}V^3}{6\pi\Gamma^2}(e^{-i\lambda_\uparrow - i\lambda_\downarrow} - 1) + \frac{(\chi_e^2 + \chi_o^2)\mathcal{T}V^3}{24\pi\Gamma^2} \sum_\sigma (e^{-i\lambda_\sigma} - 1), \end{aligned} \quad (15)$$

where $G_0 = 1/(2\pi)$ is the conductance quantum per spin orientation. In order to go over to the pure charge CGF one has to set $\lambda_\uparrow = \lambda_\downarrow = \lambda$. In this case, the result of Ref. [20] is perfectly reproduced for $h = \Delta = 0$. Moreover, it had been speculated that while the terms containing a single λ correspond to single electron tunneling events, the term with the doubled counting field is brought about by a coherent electron pair tunneling in a singlet state. Eq. (15) represents the proof of this conjecture since the term giving rise to 2λ part indeed stems from the contribution originally containing the sum $\lambda_\uparrow + \lambda_\downarrow$.

Yet another justification of the validity of (15) for arbitrary U is brought about by comparing the above result to the spin-resolved statistics of charge transfer through a Kondo impurity in the unitary limit presented in [22]. Similar to the parameter mapping for the conventional current statistics one identifies the two limits of small

and large U (Kondo regime) by

$$\phi/T_K = \chi_o/\Gamma \quad \text{and} \quad \alpha/T_K = \chi_e/\Gamma, \quad (16)$$

where T_K is the Kondo temperature and ϕ and α are Fermi liquid parameters of the Kondo fixed point [29]. The similarity of these two results can be traced back to the similarity of the corresponding Hamilton operators, which not only both contain a resonant level part but also possess analogous interaction terms.

Next, we would like to discuss the linear response (linear in V) contribution. It can easily be verified that under the conditions (11) one obtains the following CGF,

$$\begin{aligned} \ln \chi(\lambda_\sigma)_{lin} &= \\ G_0\mathcal{T}V \sum_\sigma \ln \left\{ 1 + \frac{\Gamma^2}{(\chi_c\Delta + \sigma\chi_s h)^2 + \Gamma^2} (e^{i\lambda_\sigma} - 1) \right\}. \end{aligned} \quad (17)$$

This result perfectly coincides with the conjecture of the

binomial theorem formulated in [20]. It predicts that the linear response charge transfer statistics of *any* interacting region coupled to noninteracting continua is binomial and governed by the value of the transmission coefficient at the Fermi edge. In fact, the road to the construction of the spin-resolved CGF from Eq. (26) of [20] (which contains only the charge transfer generating function) is very natural and intuitive: the logarithms with different signs in front of the magnetic field term should contain counting fields for different spin projections.

The spin current statistics can easily be recovered from the above results after transition to the charge current and spin current counting fields λ, μ via $\lambda_{\uparrow,\downarrow} = \lambda \pm \mu$. One feature of (15) is the fact that the odd cumulants of spin currents are only non-zero in finite field and for the particle-hole asymmetric case $\Delta \neq 0$. This is the precise condition for the spin flow existence in a conventional noninteracting resonant level system as well. In the linear response regime the corresponding odd order cumulants are then given by

$$\langle\langle (\delta Q_\uparrow - \delta Q_\downarrow)^{2n+1} \rangle\rangle = -\frac{2TV\Delta h}{\pi\Gamma^2} (\chi_e^2 - \chi_o^2). \quad (18)$$

The even order cumulants are non-universal but n -independent as well, so that the ratio of even/odd orders (it can be seen as a generalization of the Fano factor) is given by

$$\begin{aligned} & \frac{\langle\langle (\delta Q_\uparrow - \delta Q_\downarrow)^{2n} \rangle\rangle}{\langle\langle (\delta Q_\uparrow - \delta Q_\downarrow)^{2n+1} \rangle\rangle} = \\ & = -\frac{\Delta^2(\chi_e + \chi_o)^2 + h^2(\chi_e - \chi_o)^2}{2\Delta h(\chi_e^2 - \chi_o^2)}. \end{aligned} \quad (19)$$

Going beyond the linear response regime, we find that the most fundamental feature emerging from (15) is the existence of the *invariant cross-cumulant*,

$$\begin{aligned} \langle\langle \delta Q_\uparrow^n \delta Q_\downarrow^m \rangle\rangle &= (-i)^{n+m} \frac{\partial^{n+m}}{\partial \lambda_\uparrow^n \partial \lambda_\downarrow^m} \ln \chi(\lambda_\sigma) \\ &= (-1)^{n+m} \frac{\chi_o^2 TV^3}{6\pi\Gamma^2}, \end{aligned} \quad (20)$$

for $n, m \geq 1$. Not only is this quantity non-zero in interacting systems only, it is also independent of magnetic field strength and (up to the sign) of its orders n, m . It exists in the strong coupling Kondo case as well and is found using (16) for the parameter translation between weak and strong coupling. Despite the formally identical mathematical shapes, the origin of this phenomenon is completely different for small U and in the Kondo regime. While in the weak coupling case it signifies the beginning of the spin singlet formation, in the strong coupling limit it starts to appear as soon as it becomes possible to break up (though virtually) the Kondo spin singlet. The amplitude of these correlations grows as one approaches the strong coupling fixed point. In principle, in addition to the conventional linear conductance (which approaches the unitary limit of almost perfect conductance)

the cross-cumulant can also be regarded as a measure of how deep in the Kondo regime the system in question is being.

To conclude, we have analyzed the non-equilibrium spin resolved FCS of the Anderson impurity model by calculating the CGF of the probability distribution to transfer a fixed amount of charge with preselected spin orientation during very long waiting time interval. Our results perfectly agree with existing predictions for the statistics of charge transfer. The CGF indeed supports the interpretation that the electron transport in such a system is mediated not only by single charge tunneling but by correlated transfer of electron pairs in a singlet state. Moreover, the emerging expressions confirm the previously conjectured statistics in finite field beyond the particle-hole symmetric situation. In the linear response regime, it turns out to be binomial and to factorize in different spin channels. Finally, we have discovered a new type of correlation between the spin-up and spin-down currents: a cross-cumulant. It is universal and field independent. In our view, it has a potential to become one of the quantities to measure and control the ‘quality’ of the Kondo effect in nanostructures.

We would like to thank Hermann Grabert and Dario Bercioux for many valuable discussions. This work was supported by DFG under grant No. KO 2235/2 (Germany).

-
- [1] D. Awschalom, D. Loss, and N. Samarth, eds., *Semiconductor Spintronics and Quantum Computation* (Springer, 2002).
 - [2] A. R. Rocha, V. M. García-Suárez, S. W. Bailey, C. J. Lambert, J. Ferrer, and S. Sanvito, *Nature Materials* **4**, 335 (2005).
 - [3] Ya. Blanter and M. Büttiker, *Phys. Rep.* **336**, 1 (2000).
 - [4] Ya. M. Blanter, in *CN Summer School 2005 on Nano-Electronics*, edited by Ch. Roethig, G. Schoen, and M. Vojta (Springer, 2006), Springer Lecture Notes.
 - [5] L. S. Levitov and G. B. Lesovik, *JETP Lett.* **58**, 230 (1993).
 - [6] L. Levitov, H. Lee, and G. Lesovik, *J. Math. Phys.* **37**, 4845 (1996).
 - [7] A. Di Lorenzo and Yu. Nazarov, *Phys. Rev. Lett.* **93**, 046601 (2004).
 - [8] M. Kindermann, *Phys. Rev. B* **71**, 165332 (2005).
 - [9] T. L. Schmidt, A. O. Gogolin, and A. Komnik, *Phys. Rev. B* **75**, 235105 (2007).
 - [10] K.-I. Imura, Y. Utsumi, and T. Martin, *Phys. Rev. B* **75**, 205341 (2007).
 - [11] S. M. Cronenwett, T. H. Oosterkamp, and L. P. Kouwenhoven, *Science* **281**, 540 (1998).
 - [12] D. Goldhaber-Gordon, H. Shtrikman, D. Mahalu, D. Abusch-Magder, U. Meirav, and M. A. Kastner, *Nature* **391**, 156 (1998).
 - [13] J. Schmid, J. Weis, K. Eberl, and K. von Klitzing, *Physica B* **256-258**, 182 (1998).
 - [14] J. Reichert, R. Ochs, D. Beckmann, H. B. Weber,

- M. Mayor, and H. von Loehneysen, Phys. Rev. Lett. **88**, 176804 (2002).
- [15] P. W. Anderson, Phys. Rev. **124**, 41 (1961).
- [16] A. C. Hewson, *The Kondo problem to heavy fermions* (Cambridge University Press, 1993).
- [17] Yu. Nazarov and M. Kindermann, Eur. Phys. J. B **35**, 413 (2003).
- [18] K. Schönhammer, Phys. Rev. B **75**, 205329 (2007).
- [19] L. S. Levitov and M. Reznikov, Phys. Rev. B **70**, 115305 (2004).
- [20] A. O. Gogolin and A. Komnik, Phys. Rev. B **73**, 195301 (2006).
- [21] M. J. M. de Jong, Phys. Rev. B **54**, 8144 (1996).
- [22] A. O. Gogolin and A. Komnik, Phys. Rev. Lett. **97**, 016602 (2006).
- [23] E. M. Lifshits and L. P. Pitaevskii, *Physical Kinetics* (Pergamon press, Oxford, 1981).
- [24] K. Yamada, Prog. Theor. Phys. **53**, 970 (1975).
- [25] K. Yamada, Prog. Theor. Phys. **54**, 316 (1975).
- [26] K. Yosida and K. Yamada, Prog. Theor. Phys. **53**, 1286 (1975).
- [27] N. Kawakami and A. Okiji, J. Phys. Soc. Jap. **51**, 1145 (1982).
- [28] P. B. Vigman and A. M. Tsvelik, JETP Lett. **35**, 100 (1982).
- [29] P. Nozières, J. Low Temp. Phys. **17**, 31 (1974).